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NUCLEAR SAFETY CONSIDERATIONS IN CONTAINING A URANIUM HEXAFLUORIDE RELEASE

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September 1974

UNION CARBIDE

OAK RIDGE GASEOUS DIFFUSION PLANT
OAK RIDGE, TENNESSEE

prepared for the U.S. ATOMIC ENERGY COMMISSION
under U.S. GOVERNMENT *Contract W-7405 eng 26*

CONTENTS

	Page
INTRODUCTION	5
URANIUM HEXAFLUORIDE RELEASE CONTROL AT PLANT FACILITIES	5
NUCLEAR SAFETY	9
Moderation Effect	9
Approved Control Methods	10
Borated Water	11
Water Systems	12
Natural Uranium Hexafluoride Feed	12
DECONTAMINATION OF A LARGE URANIUM HEXAFLUORIDE RELEASE	13
CONCLUSIONS	13
REFERENCES	15
APPENDIX. CALCULATED RELEASE PARAMETERS	17

NUCLEAR SAFETY CONSIDERATIONS IN CONTAINING A URANIUM HEXAFLUORIDE RELEASE

INTRODUCTION

In response to a request from the Uranium Hexafluoride (UF_6) Cylinder Handling Committee,[1] nuclear safety considerations have been given to the possible use of water in attempting to secure a significant UF_6 release from a product cylinder. It is to be emphasized that the work of this committee and other groups involved in the study of UF_6 handling procedures, and the implementation of their recommendations, will further minimize what is considered a very remote possibility of the occurrence of a UF_6 release of the magnitude studied herein. The UF_6 cylinders evaluated were the 14-ton and the 10-ton types which have been approved for 4.5% uranium-235 enrichment and the 2-1/2-ton type approved for 5% uranium-235 enrichment.[2]

URANIUM HEXAFLUORIDE RELEASE CONTROL AT PLANT FACILITIES

An accidental release of UF_6 has always been of concern at uranium enrichment plants. Not only does the potential exist for large economic loss, but the concomitant problems of health, environment, and nuclear safety may also be created during the release situation. Visibility may be extremely poor in the immediate vicinity of a release since a voluminous and dense white fog forms almost immediately, and most certainly any emergency measures taken to cope with the release will be hampered by this fog.

It is apparent that the greatest potential for a significant UF_6 release will exist when the UF_6 is in the liquid state. The probable sequence of events following a release from a 14-ton cylinder using the Reeder model[3] are listed in Table 1. The release model assumes that a cylinder valve fails when the cylinder is horizontal with respect to the ground and with the cylinder valve either in the 6 o'clock or the 12 o'clock position. In the 6 o'clock position the postulated failure could result in the entire contents of the cylinder being forcefully ejected in about 15 min. In the 12 o'clock position, it appears that one-third of the cylinder contents would be released over a longer period, probably about 2 hours. Releases from the 10-ton and 2-1/2-ton type cylinders would be correspondingly smaller. The UF_6 release temperature is taken at 220°F.

Three ORGDP facilities have been considered as potential sites for a large UF_6 release (the K-1131 Feed and Tails Withdrawal, the K-413 Product Withdrawal, and the K-1423 Toll Enrichment Facility, see Table 2). Only in the K-1423 Toll Enrichment Facility, however, are cylinders handled routinely with the valves in the 6 o'clock position. This operation occurs during the liquid transfer of UF_6 from a 48-in. to a 30-in. cylinder. It is further noted that both the release rate which is temperature-dependent and the total amount of UF_6 released at the K-413 Product Withdrawal Facility would probably be smaller than that of the Reeder model, since

Table 1

PROBABLE SEQUENCE OF EVENTS FOLLOWING URANIUM HEXAFLUORIDE RELEASE FROM 14-TON (27,000 LB UF_6) CYLINDER

Assumptions: UF_6 is in liquid phase at 220°F. Cylinder valve breaks causing UF_6 release.*

Event	Comments	Sequential HF Release
Release occurs with valve in 6 o'clock position	All UF_6 is forcefully ejected from cylinder in about 15 min. During this time 16,000 lb UF_6 will have vaporized to the atmosphere while 11,000 lb UF_6 will have accumulated near the cylinder. The latter material may require a period of several hours to vaporize.	The vaporization of 16,000 lb UF_6 in air results in the generation of 3640 lb HF which is released to the atmosphere. Approximately 1,128,000 ft^3 of 100% relative humidity air is required to supply moisture for the UF_6 hydrolysis reaction.
Release occurs with valve in 12 o'clock position	About 7900 lb UF_6 will be released from cylinder in 2 hr. By this time the UF_6 temperature will have fallen to 147.3°F (triple point). An additional 7900 lb UF_6 will be released in another 4 hr, at which time the remaining UF_6 will probably be at subatmospheric pressure but will continue to diffuse from the cylinder at a slower rate.	The vaporization of 7900 lb UF_6 in air results in the generation of 1800 lb HF which is released to the atmosphere. Approximately 557,000 ft^3 of 100% relative humidity air is required to supply moisture for the UF_6 hydrolysis reaction.

*The Reeder Model is used to indicate the sequence of events listed.[3] Although the total quantities of UF_6 released will be smaller for the 10-ton type and 2-1/2-ton type cylinders, the initial release rates would be essentially the same since a 1-in. valve is used in each of the cylinders.

Table 2

ORGDP LIQUID URANIUM HEXAFLUORIDE HANDLING FACILITIES

Facility (Building or Room Containment Volume)	Ventilation Systems	Emergency Features*	Comments
K-1131 Feed and Tails ~500,000 ft^3	5 roof fans; 13 roof vents.	a,b,c,d,f	Cylinders handled in 12 o'clock position. Roof vents cannot be closed at present.
K-413 Product Withdrawal Room 16,000 ft^3	None.	a,c,d,f	Cylinders handled in 12 o'clock position.
K-1423 Toll Enrichment 220,000 ft^3	3 6000-cfm roof fans with gravity-close louvers. 1 250-cfm rotary blower for local vent to atmosphere. 1 275-cfm (min) centrifugal blower for pigtail vent to K-1302 stack.	a,b,c,e	Cylinders handled in 12 o'clock and in 6 o'clock positions.

*Emergency Features:
a. Remote roll-up door closure.
b. Remote ventilation system start.
c. Local UF_6 release alarm.
d. Area or central control release alarm.
e. Weathervane wind indicator.
f. Sprinkler system.

the actual cylinder temperatures at this facility are much lower than the 220°F assumed, and since only 10-ton type cylinders are used. Thus, the potential for a significant release of enriched UF_6 appears to be less in K-413 and K-1131 than in K-1423.

The Operations Division Standard Operation Procedure[4] calls for the immediate evacuation of all personnel from the affected building followed by efforts to contain and isolate the release within the building. The advantages of this policy are self-evident but are numerated below with specific comments directed to the applicable facility:

1. By denying the amount of moist air immediately available for the UF_6 hydrolysis reaction, the subsequent generation of HF may be reduced significantly. For example, in the K-1423 Facility, the quantity of HF generated during a 15-min release period could be reduced by as much as a factor of 5. (Compare in Tables 1 and 2 the building volume with the volume of moist air required for a 16,000-lb UF_6 release.)

2. The UF_6 gas, even at a temperature of 220°F, is considerably heavier than air. Thus, when confined to a building, the natural tendency of UF_6 would be to settle out and eventually hydrolyze to form uranyl fluoride. The HF generated from this slow hydrolysis would thus be more readily controllable.*

3. The K-1131 Facility will normally contain sufficient moist air to hydrolyze any anticipated UF_6 release up to 2 hours duration. Also, the building appears to be large enough to contain the release without significant building overpressure. (At present, however, there is no available mechanism to close the roof vents of the K-1131 building.)

4. The K-413 Facility volume of the product withdrawal room is so small that, if isolated, only a nominal quantity of HF would be generated by a UF_6 release.

5. Although the process buildings per se are not leaktight, some venting of the HF gas formed during a large release may be necessary. This would probably be required only during the initial 5 to 10 min of the release, or until such time as the available moisture in the air has been consumed (see Appendix).

Not included in Table 2 are two heating hoods located next to the southeast corner of the K-413 building. These hoods are used for sampling cylinders of natural UF_6 feed, with valves maintained in the 12 o'clock position. Although the heating hoods are scheduled to be phased out of operation when new equipment becomes available at K-1423,** it is noted that a UF_6 release at this location could probably not be confined within the hoods themselves.

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*Uranyl fluoride deposits resulting from this settling phenomenon have been observed on the bottom of the UF_6 gas storage tanks.

**A new autoclave has recently been placed in operation at K-1423. The autoclave, which was designed as a containment vessel, has received nuclear safety approval.[5]

NUCLEAR SAFETY

The following evaluation is considered applicable to UF_6 releases involving the 14-ton, 10-ton, and 2-1/2-ton type UF_6 product cylinders where nuclear safety is based primarily upon control of the hydrogen moderation, rather than upon cylinder dimensions, volume, or mass of uranium-235.

MODERATION EFFECT

In reviewing the nuclear safety aspects of water usage for UF_6 emergency applications, it may be helpful to look briefly at the moderation effect in criticality. Shortly after the discovery of uranium fission, it was found that good neutron moderators, such as water, greatly increased the fission probability, thus reducing the quantity of uranium required for criticality. The question of water usage in both process and emergency applications has, therefore, always been a factor of major concern wherever fissile materials are stored or handled.

The moderation effect is so great that it is almost as important for criticality, and in some cases more so, than the degree of enrichment of the fissile uranium-235 isotope itself. For example, it is well known that for uranium-235 assays of about 5% or less, uranium can not be made critical unless moderated.[6] A bench-mark experiment with seven 30-in. UF_6 cylinders was used to demonstrate this point during the development of current moderation control procedures at ORGDP, see Figure 1. While moderation is not required for criticality if the assay is greater than 5%, it can reduce the amount of uranium required by a factor of at least 100.

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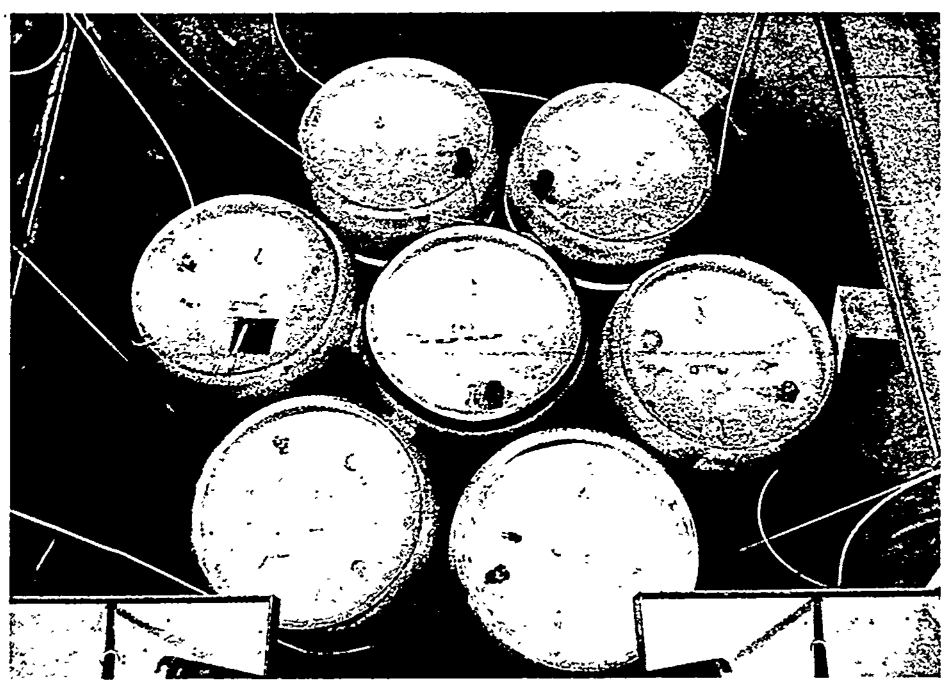

Figure 1
SEVEN 30-INCH-DIAMETER U(4.2)F6 CYLINDERS IN REFLECTOR WATER TANK

Thus, because of this important moderating property of water, relatively small amounts of uranium material can be made critical, if moderated, and the *individually-safe* units are also correspondingly small. At the 5% enrichment level of interest, for example, the safe mass of uranium is only 35.3 lb, see Table 3.

Table 3

NUCLEARLY-SAFE VARIABLES FOR THE OAK RIDGE GASEOUS DIFFUSION PLANT

Enrichment, wt % Uranium-235	Cylinder Diameter, in.	Sphere Diameter, in.	Slab Thickness, in.	Mass	
				Uranium-235, lb	Uranium, lb
≈ 90.0	5.00	8.2	1.5	0.772	-
40.0	6.00	9.2	2.0	0.904	2.27
10.0	8.20	11.8	3.6	1.320	13.20
5.0	10.25	14.6	5.0	1.760	35.30
2.0	16.00	22.3	8.7	4.410	220.00
1.0	39.60	53.4	24.0	50.000	5000.00
0.9	Infinite	Infinite	Infinite	Infinite	Infinite

Notes: 1. Units are individually safe for optimum water moderation and reflection where the uranium density of the system does not exceed 3.2 g uranium/cc.

2. All units must be spaced.

3. American National Standard ANSI N16.4-1971 now lists 1%, rather than 0.9%, uranium-235 enrichment as the maximum subcritical limit for uranium solutions.

APPROVED CONTROL METHODS

In considering the use of water in process and in emergency applications, it is evident that nuclear safety can best be assured if the uranium material is constrained by the nuclearly-safe variables given in Table 3. However, in recognition of the fact that such constraints may not always be practical or feasible under emergency conditions, the ORGDP Nuclear Safety Guide, K-1019, Revision 5[7] recommends the use of steam for UF_6 release control:

"Steam which is manually controlled may be used as a means of quickly disposing of UF_6 gas from the atmosphere in the event of a release so that appropriate action may be initiated to correct the difficulty; however, the steam nozzles should be positioned so that the steam is not sprayed directly on any system components containing moderation-limited UF_6."

The inherent factors of nuclear safety in this application are the rapid dispersal and removal of the UF_6 hydrolysis products; UO_2F_2, which tends to precipitate as a solid material over a wide area, and the HF gas which is rapidly dissipated.* This method of UF_6 release control could, in effect, be considered as a form of moderation control from the nuclear safety viewpoint, since the HF moderator is self-limiting to a certain extent.

An apparent application of the steam knockdown method of UF_6 release control would be at the K-1423 or K-1131 facilities. Here, in the event of a UF_6 release, and after building evacuation and isolation procedures have been initiated, it is visualized that the UF_6 release gases could be directed through one of the roof vents. At this control point, knockdown efforts from a number of steam nozzles could prove to be both safe and effective as a means of further UF_6 containment and subsequent collection in safe geometry equipment.

In the case of a relatively small UF_6 release, for example, one in the order of hundreds rather than thousands of pounds, attention is also directed to another potentially useful control method. A mobile exhaust unit has been designed and is now being constructed for use at the K-1423 facility.[8] The unit, which has a maximum rated capacity of 75 lb UF_6 pickup, is equipped with a 90-cfm blower, a 10-in.-ID alumina trap, and an HEPA filter. Several such units might prove useful for the control of small releases and subsequent uranium recovery throughout the plant.

BORATED WATER

Studies have shown that borated water containing 10 g boron/liter would be very effective in providing nuclear safety for a homogeneous system of uranium at uranium-235 enrichments up to 5%.[9] Accordingly, a 200-gal-capacity borated water pumper has been made available as part of the ORGDP emergency equipment for criticality control, see Figure 2. The use of this equipment in possible nuclear safety situations for the prevention of an unsafe uranium accumulation might include the following:

1. Knockdown of a UF_6 release cloud.

2. Dilution of a vessel or pit containing unknown quantities of enriched uranium.

3. Decontamination efforts following a UF_6 release incident.

*A small demonstration unit which shows the effectiveness of the steam knockdown procedure has been built and tested by the ORGDP Chemical Operations Department.

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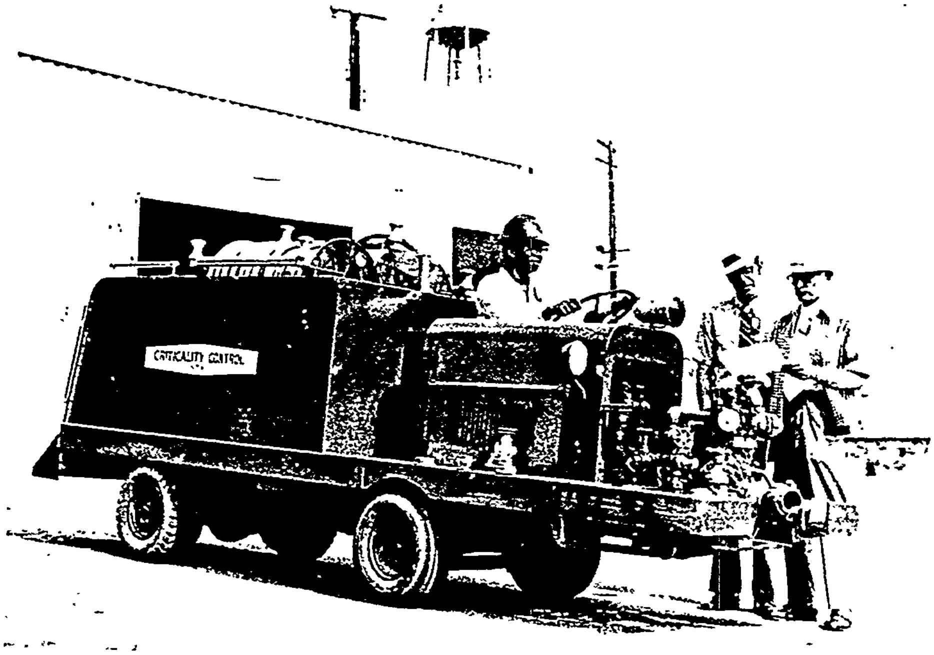

Figure 2
A 200-GALLON BORATED WATER PUMPER

However, in keeping with the ORGDP nuclear safety practice with respect to neutron absorbers, such usage would be limited to specific nuclear safety situations and under close supervisory control. Further, as in the case of the steam knockdown method, it would not be considered advisable to direct the borated water spray at any cylinder or piece of equipment containing moderation-limited UF_6.

WATER SYSTEMS

Should a UF_6 release occur in a plant facility or area where there is a distinct possibility of inadvertent mixing of water and uranium, the temporary deactivation or isolation of that water system should be considered. This would be particularly appropriate if emergency personnel were called upon to enter an area where a water release might create a potential nuclear hazard. However, such an action should be taken after due consideration of all release factors involved, and consultation with responsible personnel concerned.

NATURAL URANIUM HEXAFLUORIDE FEED

As was noted previously, two small heating hoods are located outside of the K-413 building for heating and sampling cylinders of natural UF_6 feed. Here, the use of a water spray for containment of a UF_6 release should present only minimal nuclear safety problems, since cylinders of enriched

UF_6 are not normally handled at this location. Such usage, of course, should be authorized only after first determining that the cylinder does, in fact, contain natural UF_6.

DECONTAMINATION OF A LARGE URANIUM HEXAFLUORIDE RELEASE

There has been no real experience at ORGDP to date in the decontamintion of a large release of enriched UF_6. However, as a guide to those personnel responsible for the decontamination of UF_6 release, a series of curves were prepared to delineate safe and possibly hazardous conditions for a 2200-lb release of 5% enriched UF_6 assuming various rectangular configurations, see Figure 3. For example, if the 2200-lb release were spread over a 400-ft^2 base area, the resultant thickness of the deposit would be much less than that of a safe 5-in.-thick slab. On the other hand, if the material were in a pile 12 in. high with a base area of only 9 ft^2, a possible nuclear hazard might exist. In this configuration, the material should be handled very cautiously, preferably with remote handling equipment, with the advice of a Nuclear Safety Specialist.

CONCLUSIONS

From the preceding discussion, it is evident that certain guidelines may be employed to enhance the nuclear criticality safety of any attempt to contain a significant release of 5% uranium-235 enriched UF_6. These may be summarized as follows:

1. Wherever possible the constraints of geometry and mass as defined by the established nuclearly-safe variables of Table 3 should be used to avoid a nuclear hazard.

2. Steam or borated water containing at least 10 g boron/liter may be used as a means of disposing UF_6 gas from the atmosphere. Direct contact with solid phase UF_6 or system components containing moderation-limited UF_6 should be avoided.

3. It may be desirable to isolate or deactivate any water system in a UF_6 release area where the possibility exists of moderating an unsafe accumulation of enriched uranium.

4. In those areas that handle only natural uranium, water spray may be used for containing a UF_6 release *only* after it has been determined that the material is, in fact, natural uranium.

5. Decontamination of a large release of enriched UF_6 should require the advice and consultation of a qualified Nuclear Safety Specialist.

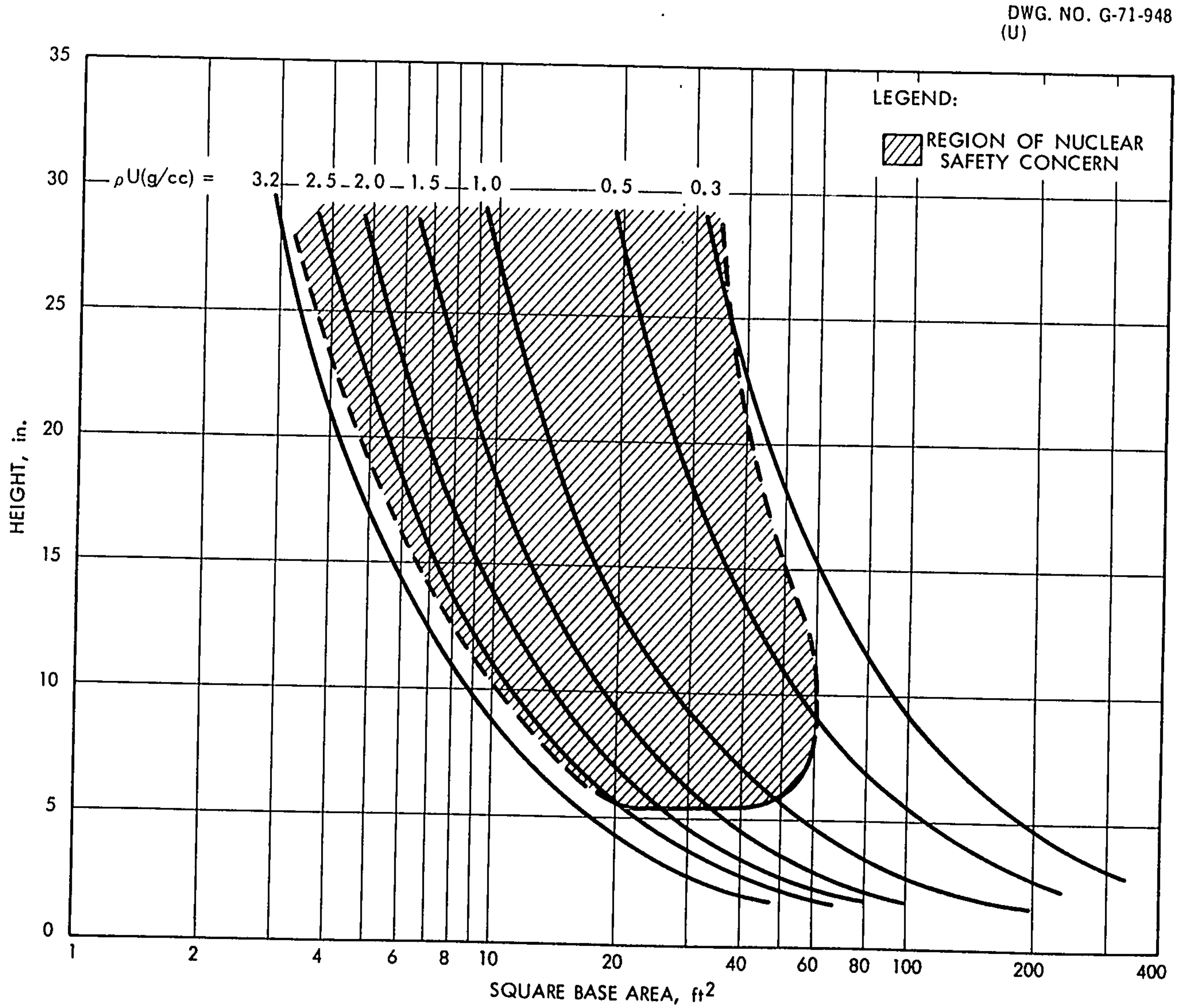

Figure 3
HEIGHT VS. SQUARE BASE AREA
2200-lb UF_6 RELEASE
5% URANIUM-235 ENRICHMENT

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APPENDIX

CALCULATED RELEASE PARAMETERS

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CALCULATED RELEASE PARAMETERS

OVERPRESSURE RESULTING FROM A LARGE URANIUM HEXAFLUORIDE RELEASE

Building codes specify that the type of structures under consideration withstand pressures of 20 lb/ft^2 (0.139 psi.), acting either inward or outward. Although the process buildings per se are not leaktight, it is apparent from the equations for overpressure developed herein that some venting of the HF gas formed during a large release may be necessary. Such venting would probably be required only during the initial 5 to 10 min of the release, or until such time as the available moisture in the air has been consumed.

The equations for overpressure were developed from standard gas laws using curve fitting techniques. Volumes correspond to the process buildings, or rooms, of interest and were assumed to contain saturated air at 77°F (100% relative humidity). UF_6 release rates of 1066 lb/min and 65.8 lb/min corresponding to the 6 o'clock and 12 o'clock models respectively, of reference 3 were used. However, in the case of the 16,000-ft^3 volume (K-413 Product Withdrawal Room), a release rate of 10.5 lb/min was assumed.

<u>K-1423 (220,000 ft^3 - 1066-lb/min release)</u>

$$OP(psi.) = 0.290\ t(min) \qquad (1)$$

After 3-5 min, the available air moisture is consumed and Equation 1 is reduced to:

$$OP(psi.) = 0.072\ t(min) \qquad (2)$$

<u>K-1131 (500,000 ft^3 - 65.8-lb/min release)</u>

$$OP(psi.) = 0.008\ t(min) \qquad (3)$$

The air volume contains sufficient moisture for complete UF_6 hydrolysis.

<u>K-413 PW Room (16,000 ft^3 - 10.5-lb/min release)</u>

$$OP(psi.) = 0.039\ t(min) \qquad (4)$$

After 18-20 min, the available air moisture is consumed and Equation 4 is reduced to:

$$OP(psi.) = 0.010\ t(min) \qquad (5)$$

STOICHIOMETRIC QUANTITIES

The UF_6 hydrolysis reaction is assumed to proceed as follows:

$$\underset{352\ \text{lb}}{UF_6} + \underset{36\ \text{lb}}{2H_2O} \longrightarrow \underset{80\ \text{lb}}{4HF} + \underset{308\ \text{lb}}{UO_2F_2} \tag{6}$$

In Figure A-1, the pounds of UF_6 reacted and the pounds of HF generated per 1000 ft^3 of moist air under varying limitations are plotted. For example, at the assumed temperature of 77°F and 100% relative humidity, 1,128,000 ft^3 of air would be required to hydrolyze 16,000 lb of UF_6. Similarly, this amount of UF_6 would generate 3660 lb of HF.

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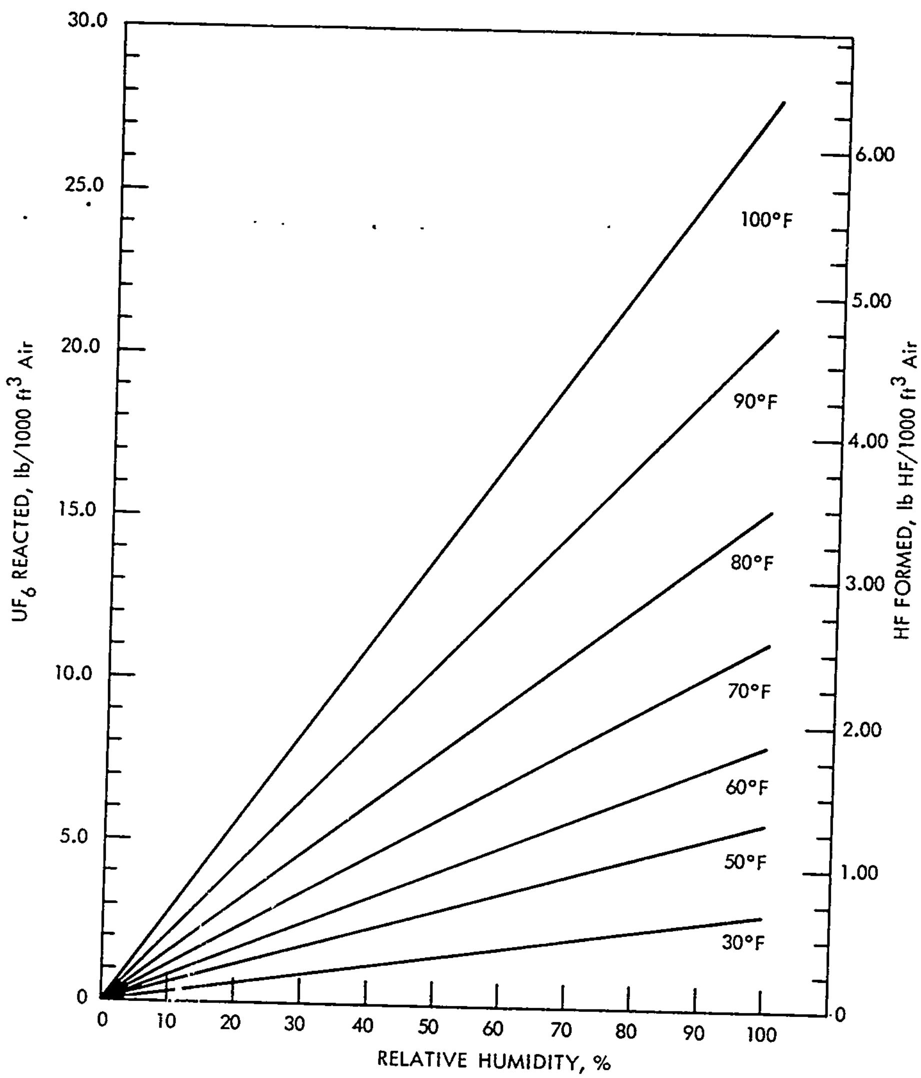

Figure A-1

REACTION OF MOIST AIR WITH URANIUM HEXAFLUORIDE